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Discussion on the Hill formula

Purpose - The purpose of the study reported in this paper was to find the correct M_t/M_∞ and Dt/a^2 data lists of Hill Formula. This paper enumerates several kinds of Hill formula data lists presented in different literatures, and we hope to draw the correct conclusion.

Design/methodology/approach - The eight different forms collected in the research project were compared. The mathematician was asked to give a suggestion from a professional point of view and to arrive at a conclusion.

Findings - In all eight cases, the third, the eighth and seventh cases are consistent and are considered reasonable and correct.

Research limitations – First-hand information was not used due to unavailability of the earliest published version of the Hill formula.

Practical implications - It is helpful to the correct application of Hill formula and the calculation of diffusion coefficient. It is also valuable to the study of dyeing kinetics.

Originality/value - The research helps to reduce and avoid the confusion in the application of Hill formula in dyeing kinetics research.

Key word - Hill formula, M_t/M_∞, Dt/a^2, dyeing kinetics

Paper type - Research paper

Introduction

Dyeing of textiles is a very important aspect in the treatment of textiles. There is a long history of mankind using dyes to dye textiles. The earliest dyes used were natural dyes extracted from plants, animals and minerals. In 1856, British chemist Sir William Perkin *FRS* made the first synthetic dye which played an instrumental role in helping the textile dyeing industry enter a new era. With the emergence of synthetic fibres and the growing need for the dyeing of such fibres, studies on the chemical structure and microstructure of fibres have promoted in-depth research on dyeing techniques and dyeing theories.

Presently, the global demand for textile is about 60 million tons. More than 80% of these textiles are dyed (Zhao, 2009). Therefore, it is important to study the dyeing technologies and dyeing theories. Dyeing theory mainly consists two parts: dyeing thermodynamics and dyeing kinetics. Dyeing thermodynamics studies whether dyes can dye the fibres and the extent to which the dyeing is likely to be achieved (dyeing balance). On the other hand, dyeing kinetics studies the speed of dyeing the fibres (dyeing rate).

It is well known that fibre dyeing process can be divided into three stages: dye diffusion from the dye bath to the fibre surface (diffusion in aqueous solution); adsorption of dye molecules by the fibre surface and dye diffusion from the fibre surface to the fibre interior (solid diffusion) (Kuroki, 1981). Due to the faster diffusion of the dye in the dyeing solution, the adsorption is almost instantaneous thus having little effect on the dyeing rate. When the dye diffuses slowly in the fibre, it has a greater influence on the dyeing rate and thus determines the dyeing rate. Therefore, diffusion in dyeing generally refers to diffusion in the fibre. The diffusion and diffusion coefficient of dye in the fibre is an important research component of dyeing kinetics. This is because the dyeing rate has important practical significance in actual production.

Data lists of Hill Formula in different literatures

The methods for determining the diffusion coefficient can be determined by the state of diffusion: unsteady state diffusion or steady state diffusion. Unsteady state diffusion can be free diffusion in an infinite system at both ends, free diffusion in a finite system at one end or free diffusion in a finite system at both ends (Kuroki, 1981). Steady state diffusion is a common method for obtaining diffusion coefficient from infinite dye bath. Crank used flake sample as their research object and solved the relationship among the dye concentration on the sample at time *t*, the equilibrium dyeing concentration and the diffusion coefficient by solving the second law equation of Fick diffusion. For fibre of radius *r*, Hill derived the relationship between the total amount of diffused matter (M_t) in *t* time in a cylinder and the amount (M_∞) of the diffusing substance entering the cylinder for an indefinite period of time and diffusion coefficient. The Hill formula is shown in Formula 1 (Fu *et al.*, 2007; Jin, 1990):

(Take in Formula 1)

In Formula 1, M is the amount of dye absorbed on the fibre; D is the diffusion coefficient; *t* is the dyeing time; V_n is the radius of the N*th* tested fibre; a is the radius of the fibre.

Because the Hill formula is an infinite series, it is more difficult to calculate and the back portion of small value can be ignored. To simplify the calculation process, M_t/M_∞ and Dt/a^2 relational tables are often used to determine the M_t/M_∞ corresponding to each Dt/a^2 value.

As long as the M_t/M_∞ is obtained, the corresponding Dt/a^2 can be obtained from the table. Since *t* is known, the fibre radius (*a*) can be measured directly using microscope or obtained by the formula $tex = \pi a^2 Ld$ (*d* is the average density of the fibre, *L* = 1000m, tex is known). Therefore, the diffusion coefficient can be obtained. However, in actual research process, we find that there are eight different versions of M_t/M_∞ and Dt/a^2 data list of Hill Formula in different literatures.

1. The first case:

The dyeing kinetics of polylactic acid fibre was researched by Fu and their colleagues (Fu *et al.*, 2007). The polylactic acid fibre was dyed with selected disperse dyes. The influence of auxiliary on diffusion coefficient *D* was determined by measuring the dye-uptake and half-dyeing time and proven by Hill equation. In this article, the data representing relationship between M_t/M_∞ and Dt/a^2 used are listed in Table I.

(Take in Table I)

2. The second case:

In the book *Dyeing and Finishing Experiment*, the diffusion coefficient of the disperse dye in the polyamide fibre was determined using the Hill formula (Jin, 1990). The data representing relationship between M_t/M_∞ and Dt/a^2 used in the above literature are listed in Table II.

(Take in Table II)

3. The third case:

The dyeing kinetics and thermodynamics of antibacterial polyester/nylon composite microfibres were studied. The dyeing properties of ordinary polyester fibre, polyester island microfiber and ordinary polyester/nylon ultrafine fibre were compared and analysed. The dyeing properties of antibacterial polyester/polyamide microfiber were discussed and the similarities and differences of dyeing performance with/from three other fibres determined. In this process, the Hill formula was applied to determine the diffusion coefficient of disperse dyes on the four types of synthetic fibres (Gao, 2007).

A series of experiments have been conducted on chemical stability of alginate fibre. Experiments on kinetics and thermodynamics of employing Lanner Red G dye to colour the alginate fibre have been carried out. These experiments used Hill formula to determine the diffusion coefficient *D* of the acid dye on the alginate fibre (Zheng, 2009).

Xu (2004) studied the dyeing capacity of bamboo fibre - a type of regenerated cellulose fibre which was evaluated in this research by contrasting it with viscose fibre and cotton fibre. A series of experiments have been conducted that proved that bamboo fibre has good dyeing performance. For dyeing thermodynamics and kinetics of bamboo fibre, this experiment used the Hill formula to determine the diffusion coefficient of direct dyes on bamboo fibres. Under the same dyeing conditions, the diffusion coefficient *D* of dyeing bamboo fibre, viscose fibre and cotton fibre in different time periods. The diffusion coefficient of bamboo fibre was higher than for the viscose and cotton fibres. The diffusion coefficient of bamboo fibre decreased with the increase of dyeing time and increased with the increase of temperature (Xu, 2004).

Purified Reactive Blue 194 was used as the target substance diffusing into the centre of viscose fibres under the condition without salt and alkali. Combined with Hill's formula, the values of diffusion coefficient of Reactive Blue 194 in the different depths of viscose fibres in cross section under the conditions of the initial dye concentration was 5g/L and the dyeing temperature was 60°C were calculated (Yang, 2020).

Disperse Red FB was employed to research dyeing thermodynamic and kinetic of cellulose acetate fibre. The kinetics parameters (diffusion coefficient D, half-dyeing time $t_{1/2}$) of cellulose acetate fibre dyed with and without dispersing agent NNO was compared (Wang, 2010).

The data representing the relationship between M_t/M_∞ and Dt/a^2, used in above-mentioned five papers, are listed in Table III.

(Take in Table III)

4. The fourth case:

The effects of ultrasonic waves on dyeing with reactive dyes on wool at low temperature and the effects of various ultrasonic wave parameters on the dyeing rate of wool were investigated. The kinetics of dyeing kinetics under the influence of ultrasonic wave and without the influence of ultrasonic wave were studied. The liquor ratio used was 1:150; which was approximated as the diffusion of the dye from the infinite dye bath to the infinitely long cylindrical fibres at both ends, in accordance with the Hill formula. It was found that the cavitation caused by the ultrasonic wave accelerated the diffusion rate of the dye in the fibre. The diffusion coefficient of the ultrasonic dyeing was 1.64 times that of the dyeing without the assistance of ultrasonic wave (Yu, 2012). In Yu's paper, the data used to represent the relationship between M_t/M_∞ and Dt/a^2 are listed in Table IV.

(Take in Table IV)

5. The fifth case:

In their process of studying the dyeing of polylactic acid fibre (PLA) by pyrazolone-type disperse dyes, the Hill formula was used to obtain the diffusion coefficient (Wang, 2014). From the calculation of the apparent diffusion coefficient of the heterocyclic disperse dye on the PLA fibre, it was found that the difference in dye structure directly affects the dye's diffusion properties in the fibre. Relevant application data are shown in Table V.

(Take in Table V)

6. The sixth case:

In the study reported in another paper, disperse dyes with high planarity and super hydrophobic structure were used to dye ultra-high molecular weight polyethylene fibre (UHMWPE) which had extreme hydrophobicity and a high level of crystallinity (Yan, 2016). The kinetic data were investigated based on Methyl Yellow and Oil Red O dyes. The results showed that the diffusion of Methyl Yellow dye in UHMWPE fibres was more efficient than that of Oil Red O dye. This indicated that the Methyl Yellow dye had a greater ability to penetrate and diffuse into the fibre. The larger the diffusion coefficient was, the faster the adsorption rate. In their paper, Hill formula was used to calculate the diffusion coefficient (Yan, 2016). The data representing the relationship between M_t/M_∞ and Dt/a^2 used by Yan (2016) are listed in Table VI.

(Take in Table VI)

7. The seventh case:

Kuroki (1981) studied the calculation of diffusion coefficient in both steady-state diffusion and non-steady-state diffusion. For an infinite system at both ends, there were two cases: diffusion in flat fibre and diffusion in cylindrical fibre. In discussing the diffusion of dyes in cylindrical fibres, the author provided data that represent the relationship between M_t/M_∞ and Dt/a^2. These data are listed in Table VII (Kuroki, 1981).

(Take in Table VII)

8. The eighth case:

Guo studied that polyamine and polycarboxylic acid dyes were applied to the dyeing of calcium alginate fibre. The effect of the main dyeing conditions on the dyeing performance and strength properties of fibre was studied. The optimum dyeing process was recommended, and the kinetic dyeing analysis of dyes was also discussed (Guo, 2016). In this article, the data representing relationship between M_t/M_∞ and Dt/a^2 used are listed in Table VIII.

(Take in Table VIII)

Results and conclusion

The same Hill formula was adopted in the above literatures, as shown in Formula 1. However, there is a deviation in the list of relationship between M_t/M_∞ and Dt/a^2 based on Hill formula. Comparison of these data showed that the data of the seventh case was largely consistent with that of the third case and the eighth case（except for the last set of date）, while the data of other cases deviate from these and each other to varying degrees. Especially, when the value of $Mt/M_\infty \times 10^2$ changed from 24 to 25, the change of D_t/a^2 became much greater. In some instances, the changes were incredible, as can be seen from Table I, Table II, Table IV and Table V. The seventh case is rooted in the Theoretical Chemistry of Dyeing as described by Kuroki Nobuhiko (1981), which quotes from the Physical Chemistry of Dyeing as described by Vickerstaff (1954). These books are considered authorities in the dyeing industry and thus it is reasonable to conclude that the third case is well detailed and reliable; and that the seventh case is nearly perfect. From the third case in this paper, it seems that the same list is used in dyeing synthetic fibre or cellulose acetate fibre with disperse dyes, dyeing viscose fibre with active dye, dyeing bamboo fibre with direct dye, dyeing the alginate fibre and calcium alginate fibre with acid dye. Dyes and dyeing conditions may not be the reason leading to the inconsistency of M_t/M_∞ and Dt/a^2 in the different literatures. We welcome contributions to further discussions from experts in this industry on this subject.

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Formula 1

$$\frac{M_t}{M_\infty} = 1 - 4\sum_{n=1}^{\infty}\frac{e^{-v_n^2 Dt/a^2}}{V_n^2}$$

$$= 1 - 4\left\{\frac{1}{5.785}e^{-5.785\,Dt/a^2} + \frac{1}{30.47}e^{-30.47\,Dt/a^2} + \frac{1}{74.89}e^{-74.89\,Dt/a^2} + \frac{1}{139}e^{-139\,Dt/a^2} + \frac{1}{222.9}e^{-222.9\,Dt/a^2} + \cdots\cdots\right\}$$

Table I Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞/ ×10^2	Dt/a^2/ ×10^2	M_t/M_∞/ ×10^2	Dt/a^2/ ×10^2	M_t/M_∞/ ×10^2	Dt/a^2/ ×10^2	M_t/M_∞/ ×10^2	Dt/a^2/ ×10^2
0	0.000	25	1.367	51	6.592	77	19.07
1	0.197	26	1.486	52	6.902	78	19.83
2	0.791	27	1.611	53	7.222	79	20.63
3	1.788	28	1.742	54	7.553	80	21.47
4	3.192	29	1.878	55	7.894	81	22.35
5	5.008	30	2.020	56	8.245	82	23.28
6	7.241	31	2.168	57	8.608	83	24.27
7	9.897	32	2.332	58	8.981	84	25.23
8	12.98	33	2.483	59	9.365	85	26.43
9	16.50	34	2.650	60	9.763	86	27.62
10	20.45	35	2.823	61	10.17	87	28.91
11	24.89	36	3.004	62	10.59	88	30.29
12	29.71	37	3.190	63	11.03	89	31.79
13	35.01	38	3.385	64	11.48	90	33.44
14	40.79	39	3.585	65	11.95	91	35.26
15	47.03	40	3.793	66	12.43	92	37.30
16	53.73	41	4.008	67	12.93	93	39.61
17	60.93	42	4.231	68	13.44	94	42.27
18	68.63	43	4.460	69	13.98	95	45.03
19	76.82	44	4.698	70	14.53	96	49.28
20	85.51	45	4.943	71	15.13	97	54.28
21	94.71	46	5.197	72	15.7	98	61.27
22	104.7	47	5.458	73	16.32	99	73.25
23	114.7	48	5.727	74	16.97	99.5	85.24
24	125.4	49	6.005	75	17.64	99.9	113.10
		50	6.292	76	18.34		

Table II Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$
0	0.000	26	1.486	52	6.902	78	19.83
1	0.197	27	1.611	53	7.222	79	20.63
2	0.791	28	1.742	54	7.553	80	21.47
3	1.788	29	1.878	55	7.894	81	22.35
4	3.192	30	2.020	56	8.245	82	23.28
5	5.008	31	2.168	57	8.608	83	24.27
6	7.241	32	2.332	58	8.981	84	25.23
7	9.897	33	2.483	59	9.365	85	26.43
8	12.98	34	2.650	60	9.763	86	27.62
9	16.50	35	2.823	61	10.17	87	28.91
10	20.45	36	3.004	62	10.59	88	30.29
11	24.89	37	3.190	63	11.03	89	31.79
12	29.71	38	3.385	64	11.48	90	33.44
13	35.01	39	3.585	65	11.95	91	35.26
14	40.79	40	3.793	66	12.43	92	37.30
15	47.03	41	4.008	67	12.93	93	39.61
16	53.73	42	4.231	68	13.44	94	42.27
17	60.93	43	4.460	69	13.98	95	45.03
18	68.63	44	4.698	70	14.53	96	49.28
19	76.82	45	4.943	71	15.13	97	54.28
20	85.51	46	5.197	72	15.7	98	61.27
21	94.71	47	5.458	73	16.32	99	73.25
22	104.7	48	5.727	74	16.97	99.5	85.24
23	114.7	49	6.005	75	17.64	99.9	113.10
24	125.4	50	6.292	76	18.34		
25	1.367	51	6.592	77	19.07		

Table III Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^2$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^2$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^2$
0	0.0000	25	1.367	51	6.592	77	19.07
1	0.1975	26	1.486	52	6.902	78	19.83
2	0.7916	27	1.611	53	7.222	79	20.63
3	1.788	28	1.742	54	7.553	80	21.47
4	3.192	29	1.878	55	7.894	81	22.35
5	5.008	30	2.020	56	8.245	82	23.28
6	7.241	31	2.168	57	8.608	83	24.27
7	9.897	32	2.332	58	8.981	84	25.23
8	12.98	33	2.483	59	9.365	85	26.43
9	16.50	34	2.650	60	9.763	86	27.62
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13	35.01	38	3.385	64	11.48	90	33.44
14	40.79	39	3.585	65	11.95	91	35.26
15	47.03	40	3.793	66	12.43	92	37.30
16	53.73	41	4.008	67	12.93	93	39.61
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23	114.7	48	5.727	74	16.97	99.5	85.24
24	125.4	49	6.005	75	17.64	99.9	113.10
		50	6.292	76	18.34		

Table IV Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞ $\times 10^2$	Dt/a^2 $\times 10^6$	M_t/M_∞ $\times 10^2$	Dt/a^2 $\times 10^4$	M_t/M_∞ $\times 10^2$	Dt/a^2 $\times 10^4$	M_t/M_∞ $\times 10^2$	Dt/a^2 $\times 10^4$
0	0.0000	26	1.486	52	6.902	78	19.83
1	0.1975	27	1.611	53	7.222	79	20.63
2	0.7916	28	1.742	54	7.553	80	21.47
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12	29.71	38	3.385	64	11.48	90	33.44
13	35.01	39	3.585	65	11.95	91	35.26
14	40.79	40	3.793	66	12.43	92	37.30
15	47.03	41	4.008	67	12.93	93	39.61
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22	104.7	48	5.727	74	16.97	99.5	85.24
23	114.7	49	6.005	75	17.64	99.9	113.10
24	125.4	50	6.292	76	18.34		
25	136.7	51	6.592	77	19.07		

Table V Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^2$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^2$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^2$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^2$
0	0.0000	25	1.367	51	6.592	77	19.07
1	0.1975	26	1.486	52	6.902	78	19.83
2	0.7916	27	1.611	53	7.222	79	20.63
3	1.788	28	1.742	54	7.553	80	21.47
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10	20.45	35	2.823	61	10.17	87	28.91
11	24.89	36	3.004	62	10.59	88	30.29
12	29.71	37	3.190	63	11.03	89	31.79
13	35.01	38	3.385	64	11.48	90	33.44
14	40.79	39	3.585	65	11.95	91	35.26
15	47.03	40	3.793	66	12.43	92	37.30
16	53.73	41	4.008	67	12.93	93	39.61
17	60.93	42	4.231	68	13.44	94	42.27
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20	85.51	45	4.943	71	15.13	97	54.28
21	94.71	46	5.197	72	15.7	98	61.27
22	104.7	47	5.458	73	16.32	99	73.25
23	114.7	48	5.727	74	16.97	99.5	85.24
24	125.4	49	6.005	75	17.64	99.9	113.10
		50	6.292	76	18.34		

Table VI Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞ /$\times10^2$	Dt/a^2 /$\times10^4$	M_t/M_∞ /$\times10^2$	Dt/a^2 /$\times10^4$	M_t/M_∞ /$\times10^2$	Dt/a^2 /$\times10^4$	M_t/M_∞ /$\times10^2$	Dt/a^2 /$\times10^4$
0	0.000	26	1.486	52	6.902	78	19.83
1	0.197	27	1.611	53	7.222	79	20.63
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3	1.788	29	1.878	55	7.894	81	22.35
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5	5.008	31	2.168	57	8.608	83	24.27
6	7.241	32	2.332	58	8.981	84	25.23
7	9.897	33	2.483	59	9.365	85	26.43
8	12.98	34	2.650	60	9.763	86	27.62
9	16.50	35	2.823	61	10.17	87	28.91
10	20.45	36	3.004	62	10.59	88	30.29
11	24.89	37	3.190	63	11.03	89	31.79
12	29.71	38	3.385	64	11.48	90	33.44
13	35.01	39	3.585	65	11.95	91	35.26
14	40.79	40	3.793	66	12.43	92	37.30
15	47.03	41	4.008	67	12.93	93	39.61
16	53.73	42	4.231	68	13.44	94	42.27
17	60.93	43	4.460	69	13.98	95	45.03
18	68.63	44	4.698	70	14.53	96	49.28
19	76.82	45	4.943	71	15.13	97	54.28
20	85.51	46	5.197	72	15.7	98	61.27
21	94.71	47	5.458	73	16.32	99	73.25
22	104.7	48	5.727	74	16.97	99.5	85.24
23	114.7	49	6.005	75	17.64	99.9	113.10
24	125.4	50	6.292	76	18.34		
25	1.367	51	6.592	77	19.07		

Table VII Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞	Dt/a^2	M_t/M_∞	Dt/a^2	M_t/M_∞	Dt/a^2
0.998	1.000	0.708	0.150	0.299	0.020
0.961	0.500	0.524	0.070	0.183	0.007
0.878	0.300	0.361	0.030	0.130	0.003

Table VIII Relationship between M_t/M_∞ and Dt/a^2

M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$	M_t/M_∞ $\times10^2$	Dt/a^2 $\times10^4$
0	0.0000	25	136.7	51	659.2	77	1907
1	0.1975	26	148.6	52	690.2	78	1983
2	0.7916	27	161.1	53	722.2	79	2063
3	1.788	28	174.2	54	755.3	80	2147
4	3.192	29	187.8	55	789.4	81	2235
5	5.008	30	202.0	56	824.5	82	2328
6	7.241	31	216.8	57	860.8	83	2427
7	9.897	32	233.2	58	898.1	84	2523
8	12.98	33	248.3	59	936.5	85	2643
9	16.50	34	265.0	60	976.3	86	2762
10	20.45	35	282.3	61	1017	87	2891
11	24.89	36	300.4	62	1059	88	3029
12	29.71	37	319.0	63	1103	89	3179
13	35.01	38	338.5	64	1148	90	3344
14	40.79	39	358.5	65	1195	91	3526
15	47.03	40	379.3	66	1243	92	3730
16	53.73	41	400.8	67	1293	93	3961
17	60.93	42	423.1	68	1344	94	4227
18	68.63	43	446.0	69	1398	95	4503
19	76.82	44	469.8	70	1453	96	4928
20	85.51	45	494.3	71	1513	97	5428
21	94.71	46	519.7	72	1570	98	6127
22	104.7	47	545.8	73	1632	99	7325
23	114.7	48	572.7	74	1697	99.5	8524
24	125.4	49	600.5	75	1764	99.9	9990
		50	629.2	76	1834		